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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric black carbon and sulfate concentrations in Northeast Greenland" *by* A. Massling et al.

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Response to reviewer 3:

1)When referring to any filter based measurements, these must be reported as EBC (equivalent black carbon , Petzold et al., 2013), along with the conversion scheme used to derive EBC from absorption coefficient. So pls replace BC by EBC everywhere in the manuscript.

Author: This was also mentioned by another reviewer and is correct. We will change this issue in the revised version of the manuscript and refer to the reference that is listed.

2) Abstract is too generic, pls include specific details, lines 7-12“A correlation between BC and sulfate: : :..”. Pls rephrase. This sentence is awkward. A strong correlation doesn’t state that transport of primary emitted BC particles is accompanied by aging: : :.A strong correlation doesn’t state that all of these processes are happening: : :.These correlations among BC and sulfate have been published before (Gong et al., 2010; Hopper et al., 1992). A strong correlation among two pollutant means they are released from the same sources or same region in case of primary source, including the similar depositions: : :.Fossil fuel combustion releases BC as primary particle and also gaseous SO2. The condensation of SO2 on to the primary particles is a very quick process and conversion to sulfate also doesn’t require a lot of time in the source regions: : :..En route from source regions to the receptor site, the aging of aerosol occurs with externally mixed aerosols converted to internally mixed aerosols. The other sources contribute to this mix, it could be biomass burning (for BC), biogenic aerosols from the ocean (DMS conversion to Sulfate and MSA): : :and during summer, it could also be biogenic from the forest.

Author: A similar statement was also discussed by another reviewer, we will revise this section. Please see that the manuscript contains a section that explains why aging of BC by sulfate is reasonable and discusses the air mass transport with low wet deposition over ice. DMS is not likely to be a main contributor here, as the air masses mostly pass over ice-covered surfaces as discussed in the manuscript. Fossil fuel combustion from Europe does not include much SO2 except from shipping and aviation. Finally SO2 has an average lifetime of 1 week but could be much longer during the Arctic winter and spring where there is little or no sun light. In fact SO2 concentration is building up during winter and after the polar sunrise it decreases and sulfate is building up (e.g. Nguyen et al. 2013, Heidam et al. 2004). The correlation between sulfate and BC is thus a dependency on source area and the fact that BC particles must grow into a size with minimum deposition velocity, see answer to reviewer 2. All together this is why we say that BC and sulfate have only partly common sources. Also in the case that they have common sources, the sulfate will lead to a substantial diameter growth of the BC

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particles leading to the proposed mechanisms. This will be added and discussed in more detail in the revised version of the manuscript.

3) Lines 22-24 pls delete this sentence: : :"during winter and spring the Arctic atmso-here known to be impacted by: : :.." .This is a known fact and should not be in the abstract: : :

Author: Yes, we agree on this statement and we will delete it in the revised version of the manuscript.

4) lines 25-26, Pls add flaring is also recognized as an important source (Stohl et al., 2013)

Author: Thanks for that important comment. Flaring will be mentioned in the revised version of the manuscript including the reference listed here.

5) pg 11468 lines 1-4, What is the source of anthropogenic sulfate during the summer? Usually since the frequent transport from the south is slow down, sulfate is usually associated with the conversions of biogenically produced dimethyl sulfide emissions. Is the data screened for local contamination from the diesel generators?

Author: We have previously had two years of parallel filter pack measurements at either site of Station (Long wave hut LW and Flygers Hut). There was an excellent agreement between the two series of measurements for sulfate. The slope was close to one with R2 = 0.9739 (Goodsite et al. 2014). Thus on the timescale of one week we cannot see the effect of Station Nord. Flygers Hut is located 2.5 km south of the Station. The station is manned year round with 6 soldiers and the visitors so the source strength is remote and rather small. NOx concentrations have sometimes very sharp concentration peaks which most likely are caused by local sources but as previously stated the effect of the Station on sulfate is negligible. DMS emitted during summer is very likely an important source but not during winter.

6) line 8, pls add Sharma et al., 2013.

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Author: Thanks for that addition. This reference will be added in the revised version of the manuscript.

7) line 21-23, pls add Petzold et al., 2013 and use new recommendations as per this manuscript. Replacing BC with EBC (equivalent black carbon as previously suggested).

Author: Thanks. As mentioned above, this reference will be added in the revised version of the manuscript.

8) lines 23-25, " : : :.is put into context of possible aging and transport: : :." It is extremely important to mention the same source region such fossil fuel: : :.possible aging and transport mechanisms: : :.

Author: As mentioned in the comment above, we will add these facts and also discuss same sources in the revised version of the manuscript.

9) pg 11469, lines 5-10, EC/OC was collected at military station 2.5 km from the main sampling station. MAAP and sulfate were located at the main site hut 2.5 km southeast of the military camp. What is the predominant wind direction? Are the filter data sectored to stop sampling when the winds are from southeast sector? Does the weekly MAAP data screened out for military camp contamination? Pls comment.

Author: The predominant wind direction at the camp is from south west. The filter pack is not stopped for specific wind directions but as discussed above, the sampling site is about 2.5 km away from the main camp. For the MAAP we used daily data, which also cannot be screened for short time pollution as this does happen on a minute or hour base, when very rarely a truck passes by at this remote site. The data used in this article may have some minor contribution from local sources which cannot be fully excluded. Based on the fact that sulfate and BC are measured remotely out of the camp we assume that this pollution is only minor and on very short time scales as was figured out for earlier datasets when NOx was also measured at the station.

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10) lines 19-20, pls specify why used 6.6 m2g-1 for conversion of light absorption of MAAP measurements to EBC mass and uncertainty associated with the measurement in the abstract where mention the MAAP measurements.

Author: Yes, we will make this more clear and refer to the right reference and state why this factor is used in the revised version of the mansucript. We also mentioned this in another comment above.

11) lines 22-25, pls replace uncertainty in black carbon to uncertainty in the light absorption coefficient measurements. Also suggest, uncertainty in equivalent black carbon could be as large as a factor of 2.

Author: You are right. This is more pointing out the uncertainty that is related to the parameter of light absorption coefficient. We will also add your suggestion, that the uncertainty in EBC can be as large as a factor of 2 in the revised version of the manuscript.

12) p 11473 line 21, : : :"There are: : :: : :sulfate and BC which is large and cannot be estimated for this study." line 22-25, pls rephrase, "The anthropogenic emissions of SO2 and BC: : :" to " the annual averaged emissions of SO2 and BC were used in the model, not taken into account the seasonal variability in these emissions which could be about 20%". Is this number correct? How did you get seasonal variation in BC emissions? These are usually listed as annually averaged values in the emissions database: : :

Author: The text at p 11473 line 21 will be changed: Old text: There are several uncertainties connected to the model calculations of sulfate and BC. New text: There are several uncertainties connected to the model calculations of sulfate and BC which are large and cannot be estimated for this study.

Author: The text at p 11473 line 22-25 will also be rephrased. The mentioned uncertainty in 20% is not for the seasonal variability but for the yearly average. It is probably at least 20% and is perhaps even larger. 20% is the uncertainty for the yearly aver-

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age of Denmark. It will be mentioned in the revised version of the manuscript how the seasonal variation of BC is done due to a comment from another reviewer. Old text: The anthropogenic emissions of SO2 and BC are given as total yearly values, which have an uncertainty of 20% or more. New text: The annual averaged anthropogenic emissions of SO2 and BC were used in the model and these could have an uncertainty of at least 20 % on the yearly basis.

13)pg 11476, lines 24-30, pls cite Sharma et al., 2013 for seasonal pattern at Alert and Zepplin Stations.

Author: Thank you for that information. This reference will be added in the revised version of the manuscript.

14) lines 6-10 all analytical systems have a detection limit for various components, pls determine the detection limits for MAAP and Thermo-optico method. How different are these measurements from detection limits especially during summer? Line 18 Pls replace "Minimum sulfate concentrations were close to zero: : :." With the detection limit value: : :..

Author: There will be a more detailed quantification on these parameters in the revised version of the manuscript, so that it also gets clear how valuable the data are during the low concentration periods as e.g. in summer.

15) pg 11477 lines11-19, The lower ratio in the summer of sulfate to EC could due to the fact that lower sulfate is present due to only biogenic impact and higher EC due to biomass burning. Do you also have measurements of potassium due to biomass burning to back up this statement: : :

Author: From receptor analysis using PMF and COPREM we failed to identify the contribution from biomass burning (Nguyen et al. 2013, ACP). From other high Arctic stations we expect that biomass contribution is significant and we have ongoing work to distinguish more efficiently between the various sources. We have thus measured

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the chemical composition of particles with a Soot Particle Time-of-Flight Aerosol Mass Spectrometer (SP-ToF-AMS) this spring at Villum Research Station.

16) pg 11479 Line 20 replace “dimethanesulfide” by “dimethyl sulfide”. Lines19-21 “the results suggested that photo-oxidation of DMS: : :.” This is true only in the winter/spring time (30% is contributed from biogenic produced sulfate (Norman et al., 2002)). Summer time this fraction could be higher. Pls rephrase this statement.

Author: Thank you for this valuable input. We will rephrase this statement according to the listed reference in the revised version of the manuscript.

17) lines 21-24, BC and SO2 are released from the same fossil fuel sources and are long range transported. The conversion process occurs en-route from source regions to the receptor site. In lines, 304-307, you mentioned that Siberian smelters and other long-range transported anthropogenic pollution contribute to both. In the end, everything gets internally mixed as there is a long transport time from Siberia to Station Nord.

Author: Yes, this is right. Depending on the air mass transport pathways, the aerosol will have different origins and thus also different chemical composition. We expect the observed aerosol at VRS to be a complex mixture of aerosols of different origin.

18) p 11480, Lines 6-12 How are BC and sulfate partly related to same combustion sources? Your hypothesis of sulfate particles as being transport containers is kind of misleading?? This is only true if the particles are internally mixed. If you consider sulfur dioxide and then sulfate coated black carbon shells coming from fossil fuel combustion source regions, the release and mixing of particles from the metal smelters will condense on the already existing black carbon particles coming from other source regions: : :should that not be black carbon being containers for sulfate from smelters by condensing sulfur dioxide??

Author: This is rather more how you formulate it. The general outcome is the same.

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The two components mix and this mixture favors their transport to the North. EBC containing particles however, need to grow, otherwise they cannot be long range transported, see previous discussion.

lines 14-22, during the summer, sulfate should have a large proportion of biogenic contribution. Anthropogenic influence should be minimal during the summer as clearly seen in the measurement at Alert, Nunavut location. Pls separate the Nov to May period from June to October in Figure 4 (a) and (b). The regression should fall apart during the summer as also clearly evident by the data at the lower end of the curve. If there is correspondence, it might be due to local influences. Also summertime increases in the precipitation in the source region and precipitation also resulted in lower sulfate and EBC. Is the deposition of the two a selective process as sulfate being much more soluble than EBC?

Author: The relative contribution of DMS to sulfate during summer is most likely high but the absolute concentration is very low at VRS and therefore does not perturb the general correlation with EBC. We have looked into several correlations for sulfate and EBC earlier dividing the dataset into smaller time periods as e.g. summer and winter, but the total amount of data does not allow statistics that can be interpreted then further. We found that any evaluation on this is not reasonable.

19)pg 11482, lines 13-17, The regression plots of measurements of sulfate and EBC give a slope that is a factor of 4 lower than regression plots of model outputs for sulfate and EBC. Model output is overestimating sulfate and underestimating BC. That's the reason the slopes are so different with respect to measurements?? Pls include that.

Author: Yes, we think that an overestimation of sulfate and underestimation of BC is responsible for that extreme difference in the slopes for the two correlations. Reasons are also given for that high uncertainty in the manuscript and are related to corresponding uncertainties in the emissions databases.

20) The results from two black carbon techniques were compared although the two

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instruments were situated in laboratories 2 km apart and not having a common inlet.

Author: This is right and also clearly stated in the manuscript. There is also now additional information on what the reason is for the different measurement locations.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 11465, 2015.